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S681 SB

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B1X
Selected US specifications from IPC sub-class B01D

(54) **Apparatus for the desulphurisation and denitration of exhaust gases by electron irradiation**

(57) Apparatus for the desulphurisation and denitration of exhaust gases through electron irradiation of the exhaust gases, to which ammonia has been added before irradiation, comprises an exhaust gas channel and a minimum of one low energy electron beam source with an electron beam potential of 150 - 300 keV. The electron beam source is arranged con-centrically and coaxially in the exhaust gas channel and has a minimum of two, but preferably four or more electron emission apertures.

The drawing(s) originally filed was (were) informal and the print here reproduced is taken from a later filed formal copy.

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FIG.1

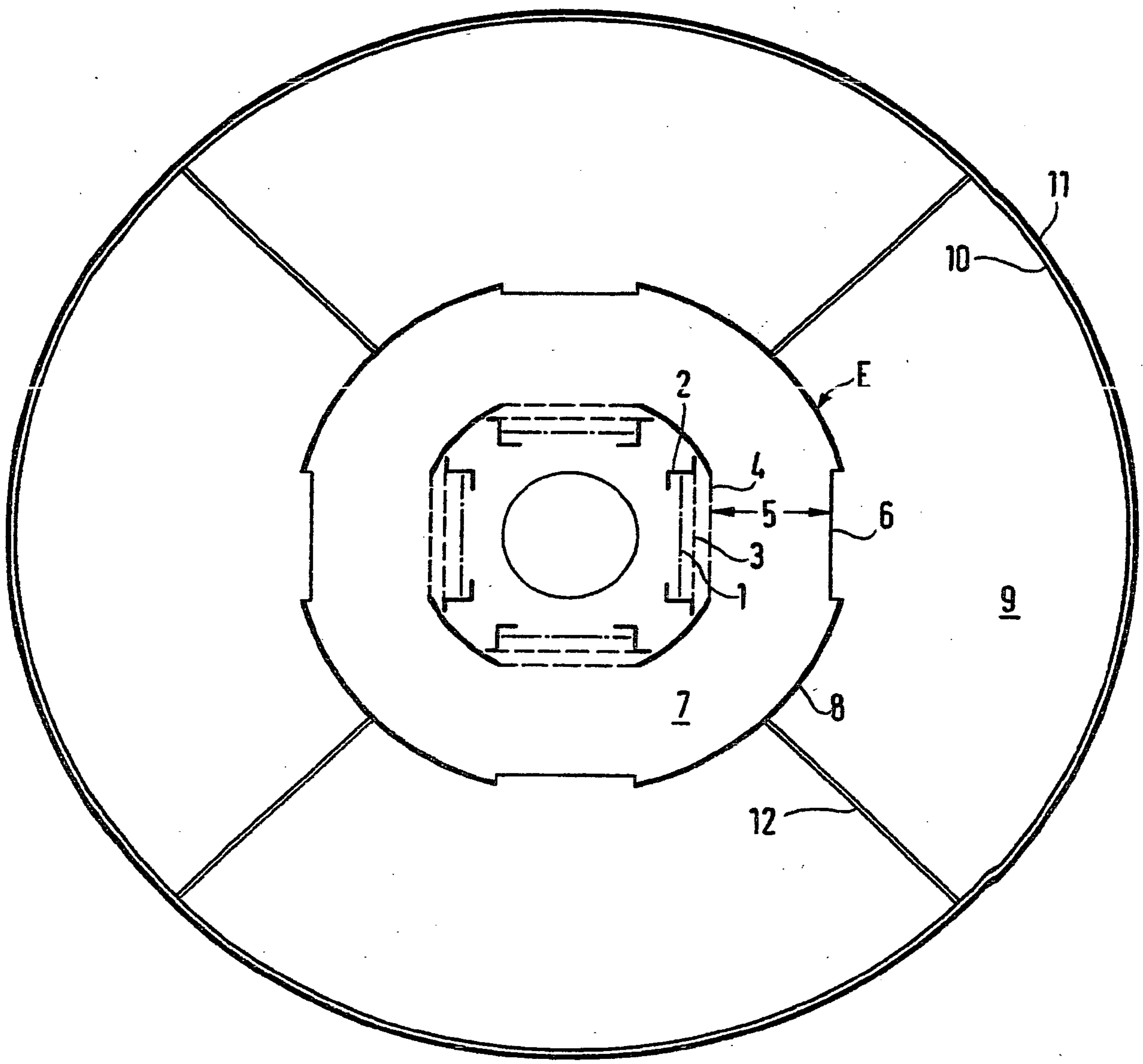

FIG.2

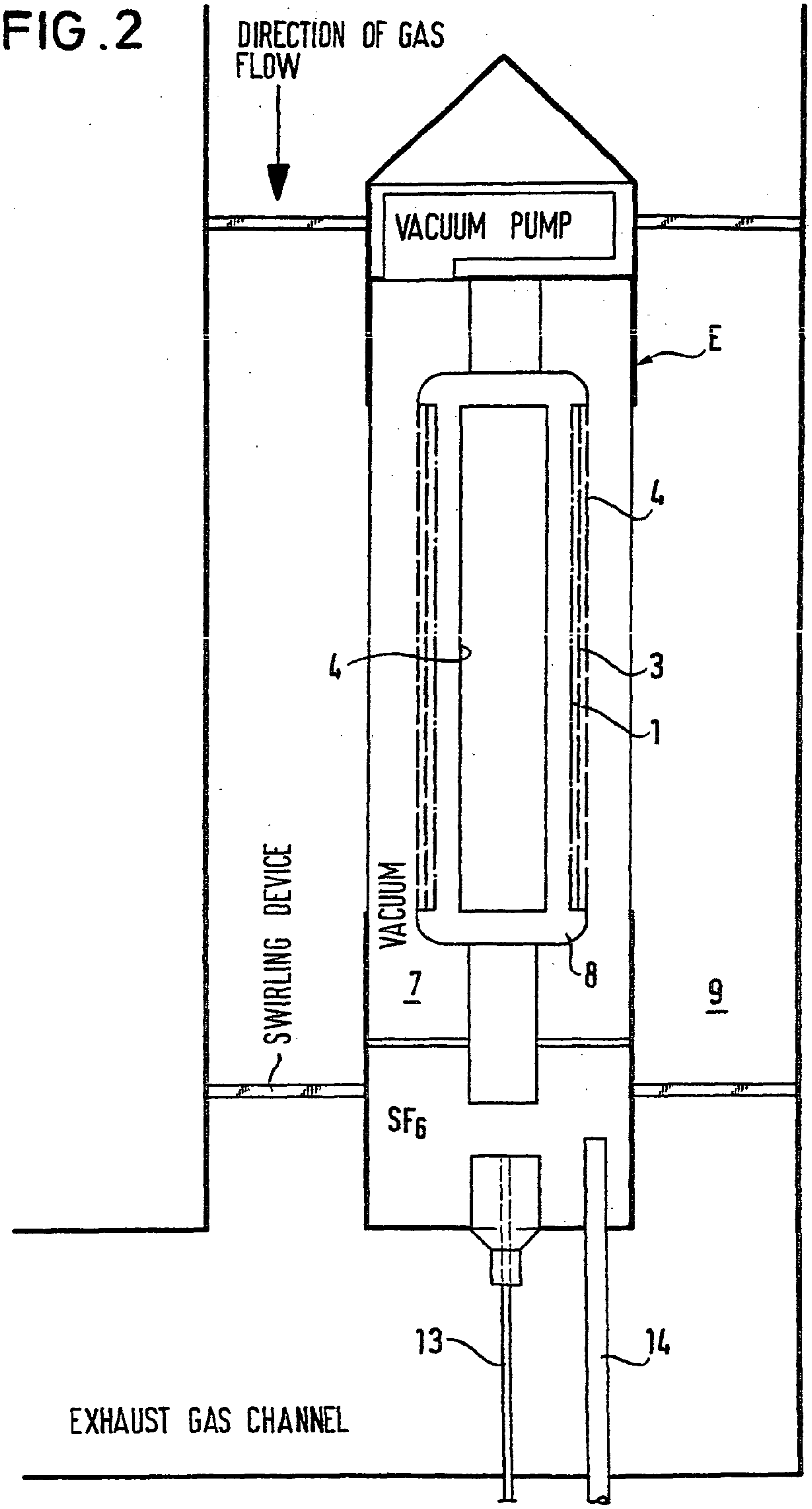
DIRECTION OF GAS FLOW
VACUUM PUMP
E
4
4
3
1
SWIRLING DEVICE
VACUUM
7
8
9
SF6
13
14
EXHAUST GAS CHANNEL

FIG. 3

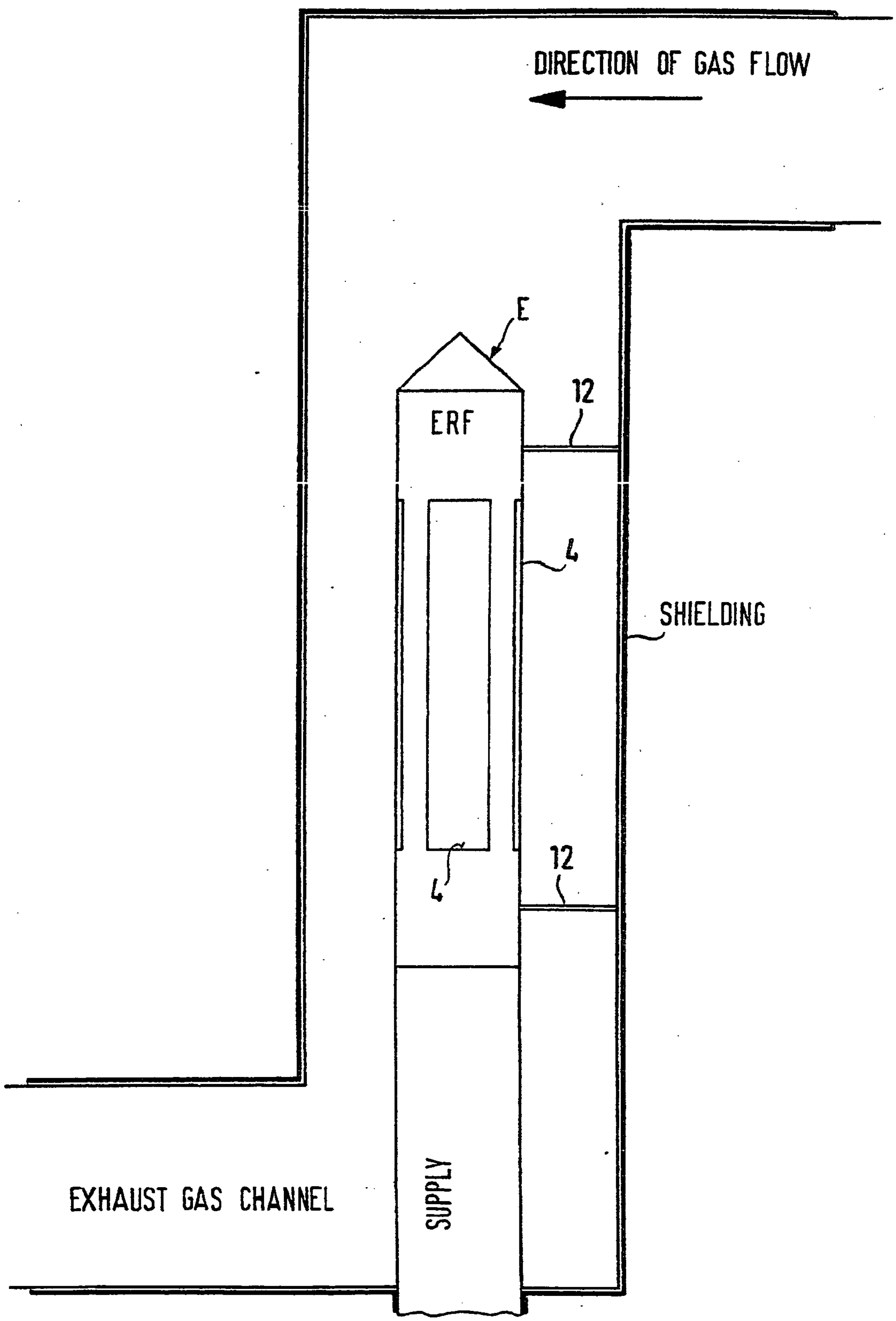
DIRECTION OF GAS FLOW
E
12
ERF
4
SHIELDING
12
4
EXHAUST GAS CHANNEL
SUPPLY

FIG. 4

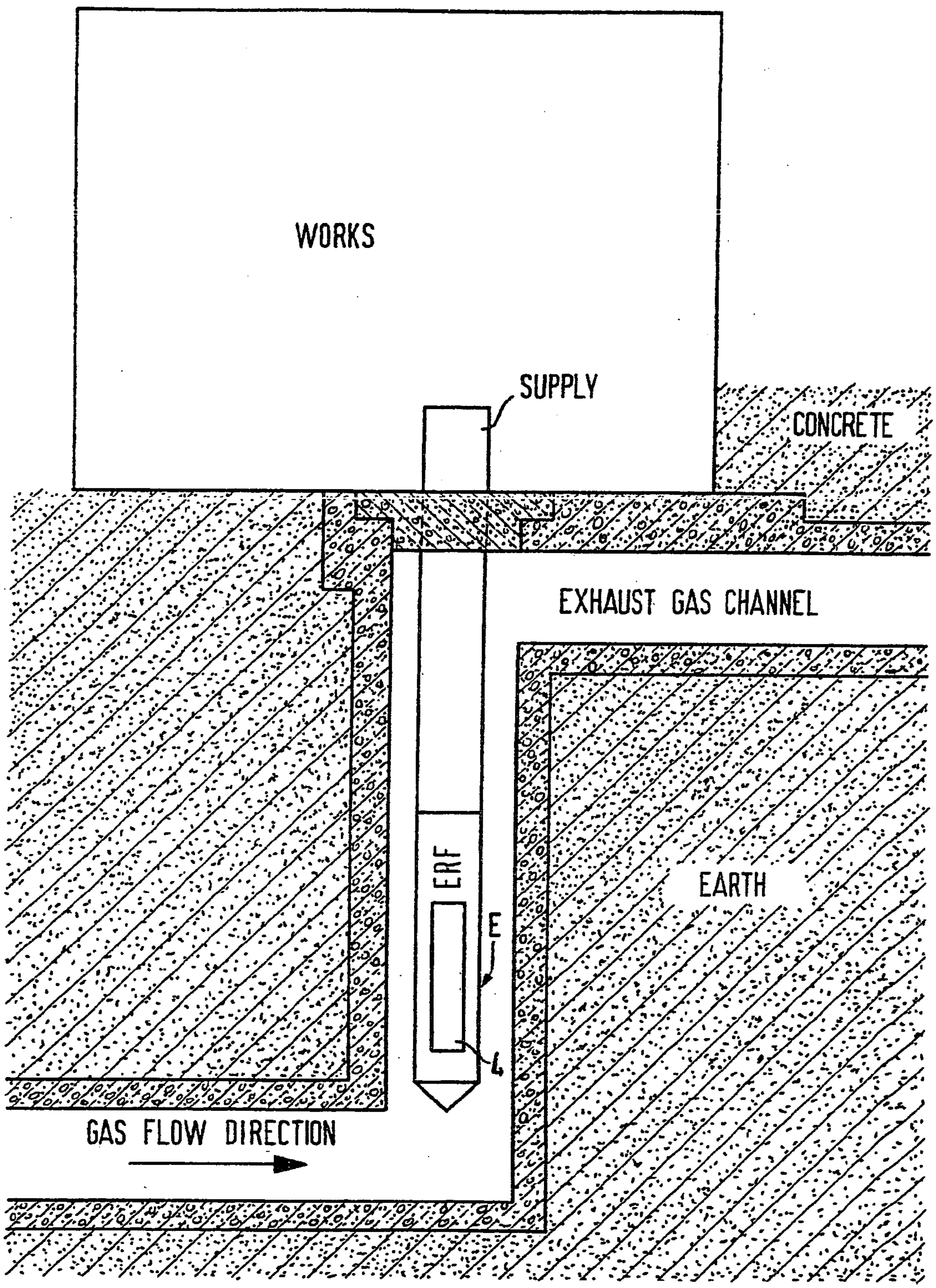
WORKS
SUPPLY
CONCRETE
EXHAUST GAS CHANNEL
ERF
E
4
EARTH
GAS FLOW DIRECTION

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FIG. 5

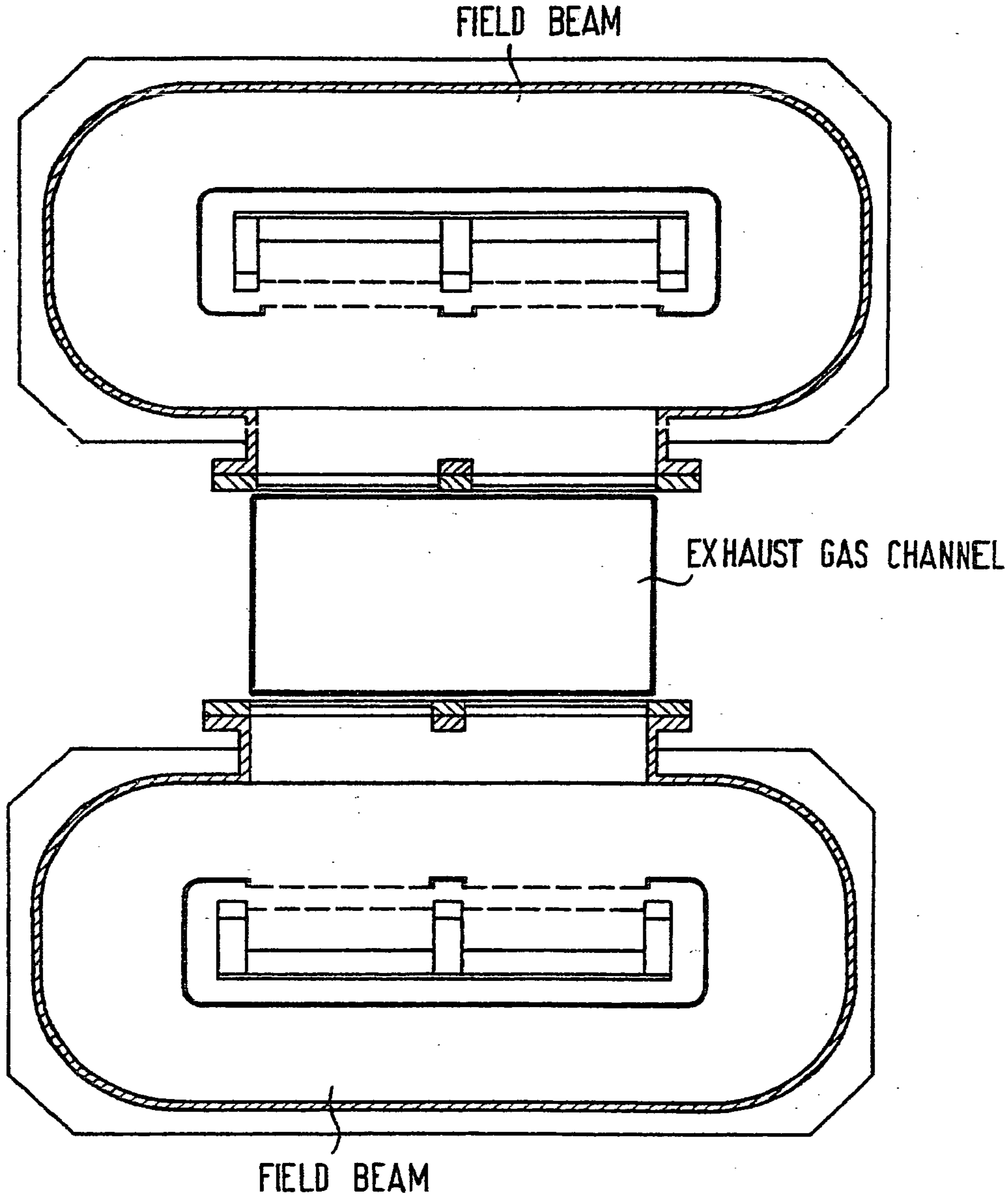

FIG.6

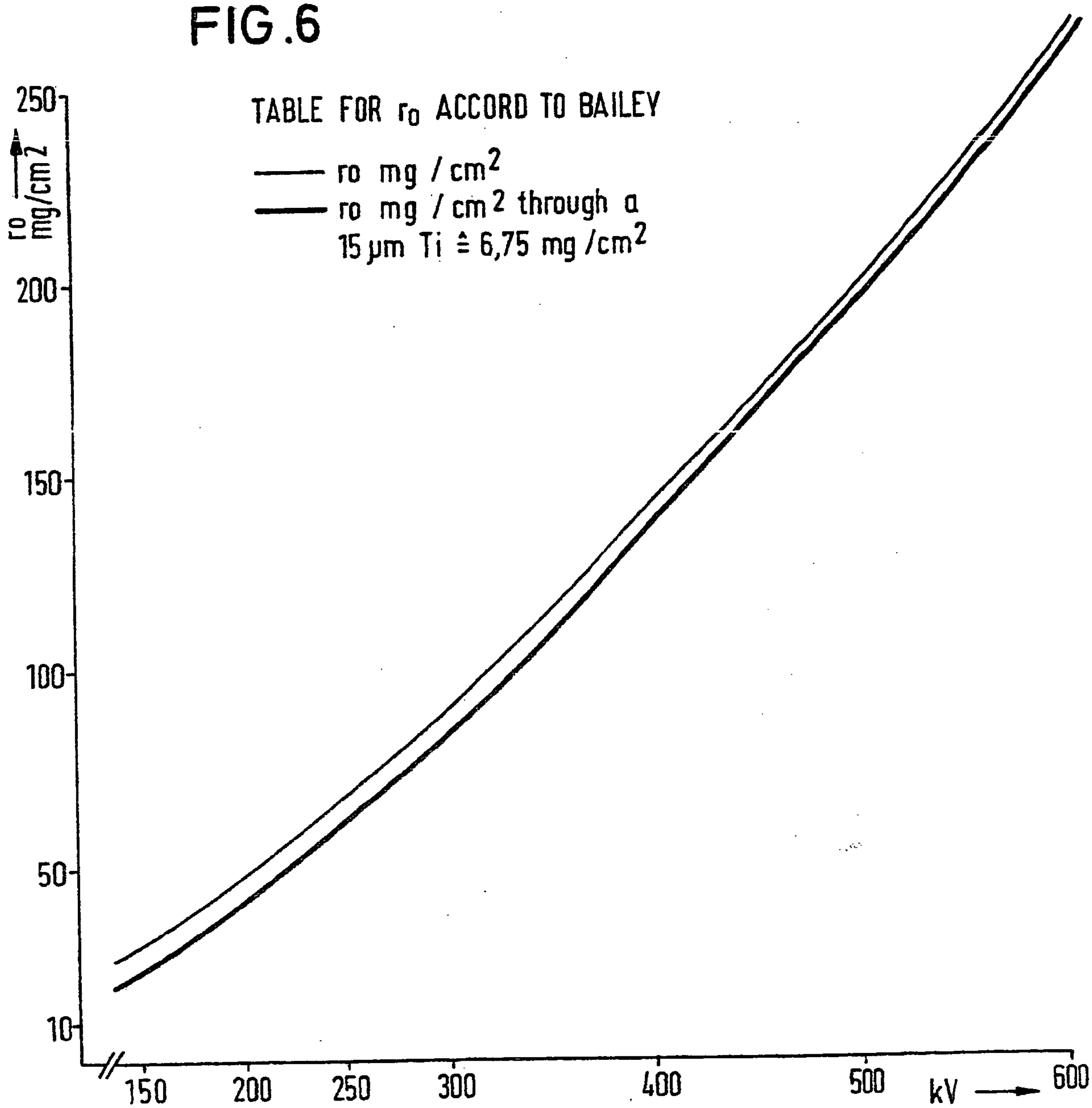
TABLE FOR r_0 ACCORD TO BAILEY
ro mg / cm2
ro mg / cm2 through a
15 µm Ti ≙ 6,75 mg /cm2
ro mg/cm2
250
200
150
100
50
10
150
200
250
300
400
500
600
kV

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FIG.7

MAXIMUM IONISATION AT 250 keV

TWO-SIDED IRRADIATION

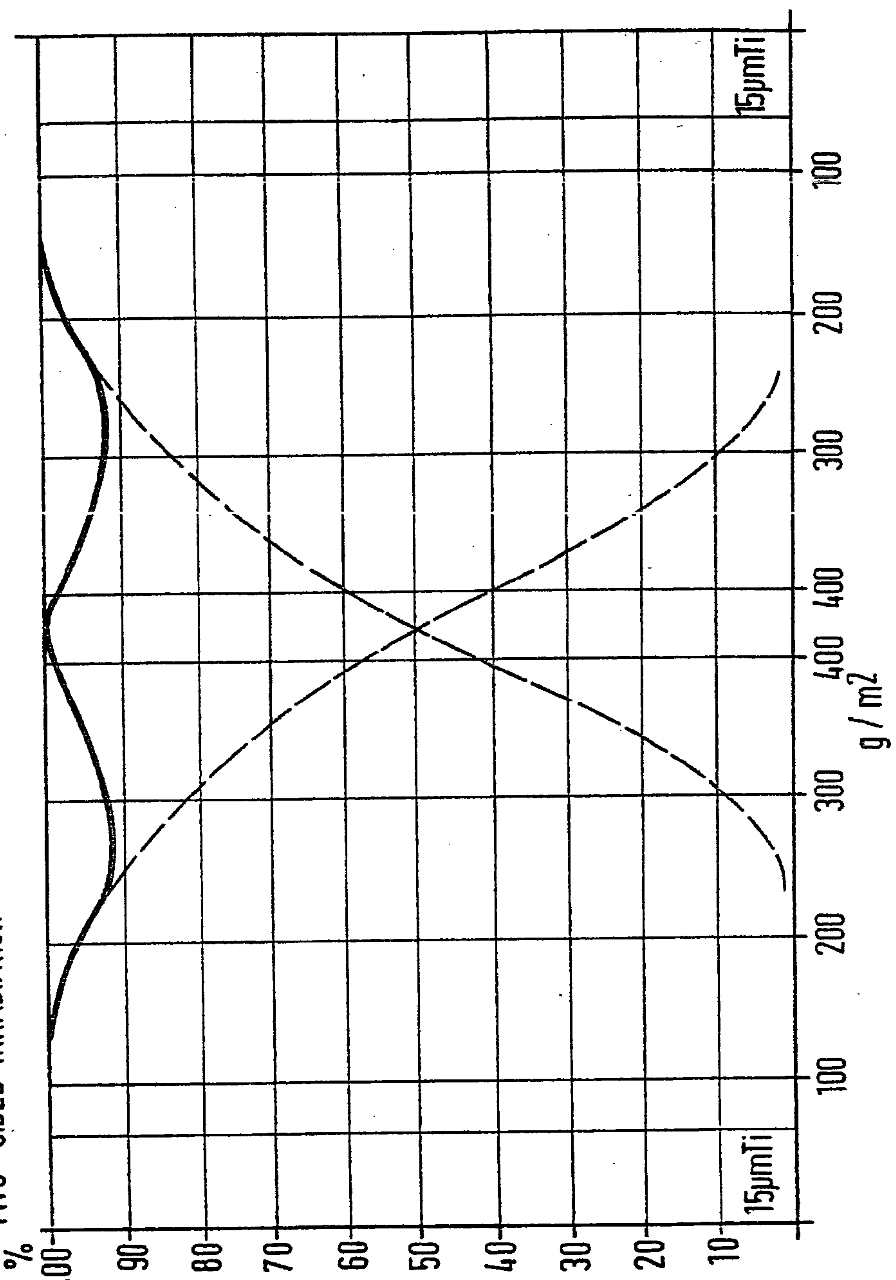

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FIG. 8

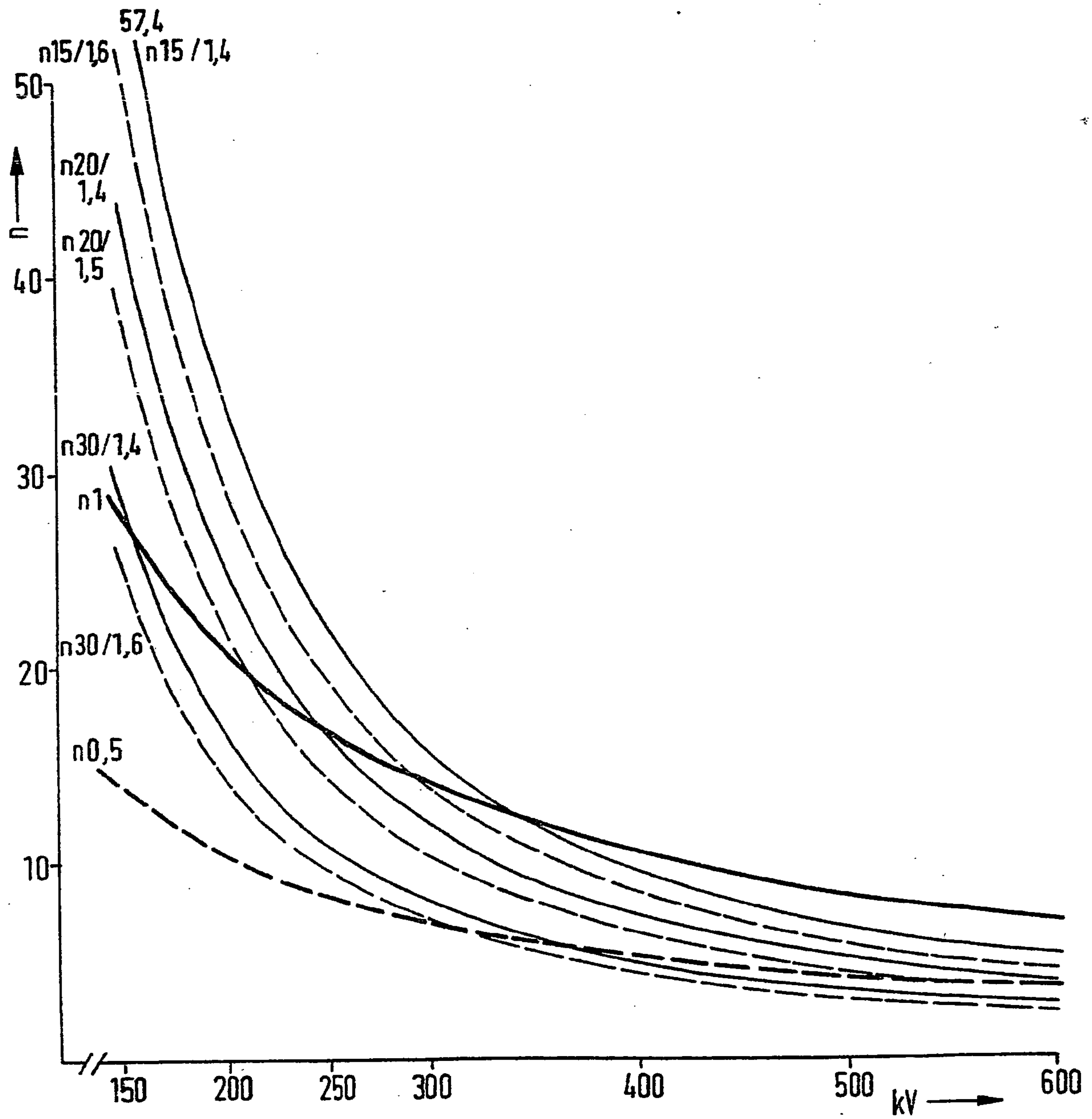
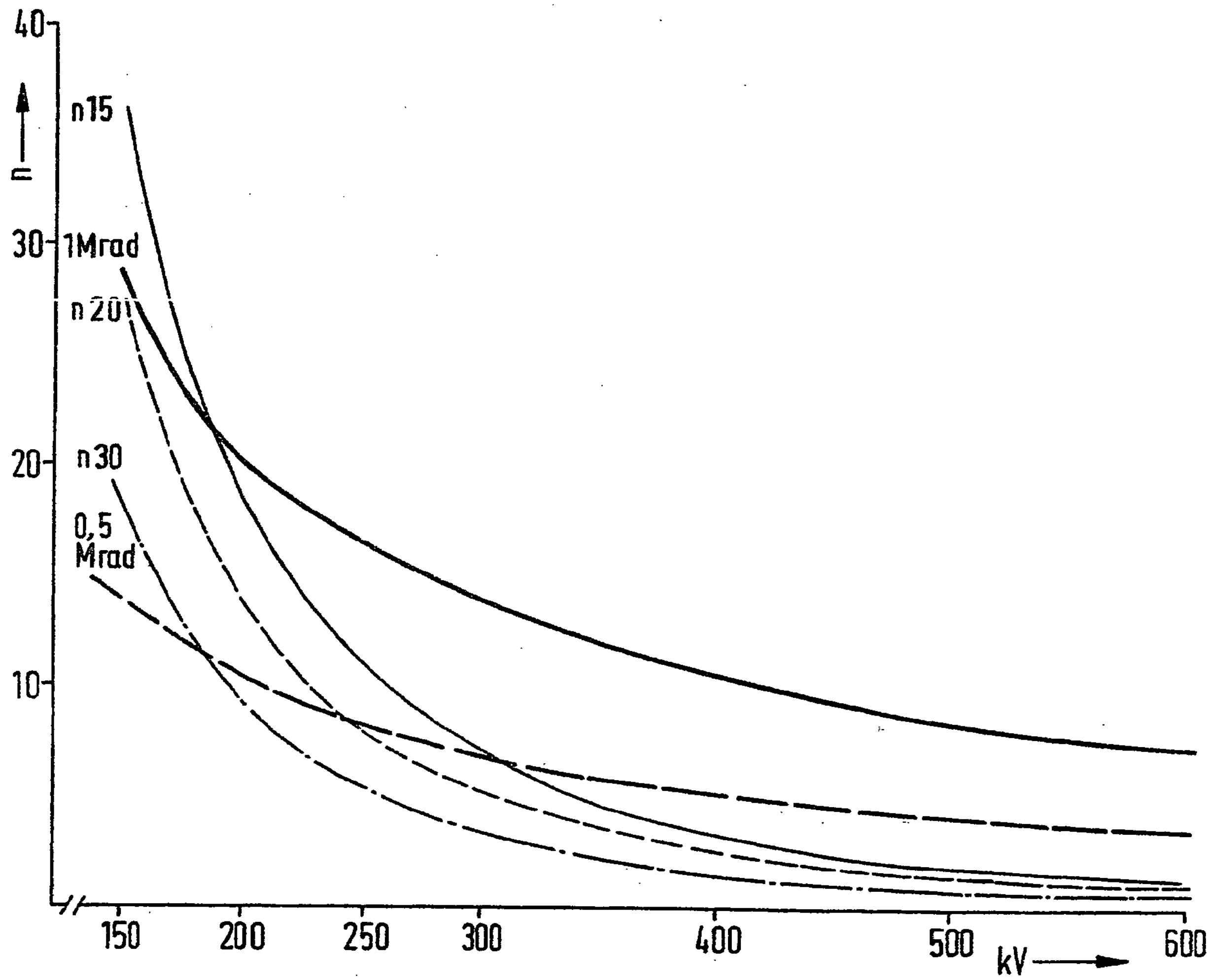


FIG. 9


40
30
20
10
n
n15
1Mrad
n20
n30
0,5
Mrad
150
200
250
300
400
500
600
kV

SPECIFICATION

Apparatus for the desulphurisation and denitration of exhaust gases by electron irradiation

The present invention relates to apparatus for the desulphurisation and denitration of exhaust gases by electron irradiation of the exhaust gases to which ammonia has been added before irradiation, the apparatus consisting essentially of an exhaust gas channel and at least one low energy electron beam source with an electron beam potential in the region of 250 keV.

The desulphurisation and denitration of exhaust gases from large scale furnaces today plays an important part in the waste management of our environment.

In addition to catalytic drying processes and a number of wet processes, which are partly simultaneous and partly selective, a physical process has been developed in Japan in recent years, in which the conversion of SO_2 and NO_x is carried out through irradiation with accelerated electrons in the presence of ammonia. This produces ammonium sulphate and ammonium nitrate which are removed by means of air filters. In this process, described for example in *Radiat. Phys. Chem.*, vol 18, nos. 1–2, pp 389–398, 1981, the exhaust gases are irradiated by two relatively high potential electron beam sources (750 keV) opposite each other in a round, continuous flow reactor with simultaneous mixing. For various reasons the use of these high beam potentials has proved to be disadvantageous.

Thus it was proposed by the applicant to carry out the desulphurisation and denitration of exhaust gases through irradiation with low energy electrons (German Patent Application P 34 03 726.8), although with this process and the device used for it, only relatively small amounts of exhaust gas can be treated, as the electron beams used are only equipped with one point cathode and electron beam deflector (scanning principle), and due to the limited electron emission from the point cathode the required high performance cannot be achieved.

In order to overcome this problem, the applicant proposed the use of low energy electron beams which achieve the required high performance due to the fact that at least two large area cathode systems are arranged in parallel in a vacuum casing, with each large area cathode system having its own electron emission aperture of the same length and width as the large area cathode system. In this device the beam potential, the electron flow, electron emission aperture loads, penetration depth of the electrons and consequently the cross-section of the exhaust gas channel are coordinated in such a way that with two electron beams situated opposite each other optimum irradiation conditions can be attained. However, for irradiation to take place, two fully independent electron beam devices, are required which are applied to an exhaust gas channel from *outside*, something seen as a disadvantage. Furthermore the exhaust gas channel and beams have to be shielded by lead sheet. Shielding in the transition electron beam/exhaust gas channel is cost intensive as the electron beams have to be taken away when maintenance work is carried out on the exhaust gas channel. This device is described in the applicant's German Patent application P 34 39 190.8.

The object of the present invention is therefore to provide a device for the desulphurisation and denitration of exhaust gases through electron irradiation of the exhaust gases, in which the use of only one low energy electron beam source is necessary.

The invention accordingly provides apparatus for the desulphurisation and denitration of exhaust gases by electron irradiation of the exhaust gases to which ammonia has been added before irradiation, comprising an exhaust gas channel and a minimum of one low energy electron beam source with an electron beam potential of 150–300 keV, in which the electron beam source is arranged concentrically and coaxially in the exhaust gas channel and has at least two electron emission apertures.

Embodiments of the present invention will now be described, by way of example only, with reference to the drawings, in which:

Figure 1 is a cross section through an exhaust gas channel with electron radial area beam;

Figure 2 is a longitudinal section through the radial area beam and exhaust gas channel;

Figure 3 shows an exhaust gas channel "horizontal" with radial area beam and lead sheet shielding;

Figure 4 shows an exhaust gas channel "vertical" with radial area beam and earth as shielding;

Figure 5 illustrates the arrangements of electron area beams and shielding during opposing irradiation;

Figure 6 is a graph showing maximum extent r_o of electron beams as a function of beam potential (according to Bailey);

Figure 7 shows ionisation curve for various beam potentials;

Figure 8 is a graphical comparison of the number of electron area beams arranged opposite each other with the number of exhaust gas channels;

Figure 9 is a graphical comparison of the number of radial ar a beams and number of exhaust gas channels.

In the drawings the reference numbers refer to the following:

E	Radial area beam as electron beam source
1	Field cathode
2	Cathode bearer
3	Extraction grid
4	Acceleration grid

5 Acceleration distance
6 Electron emission aperture
7 Vacuum
8 Side of recipient
9 Irradiation area in the exhaust gas channel
10 Outer wall of the exhaust gas channel
11 Shield
12 Suspension of beam
13 Energy supply line
14 Vacuum supply pipe

The costs for the electron beams and the shielding devices is greatly reduced by the present invention, as instead of area beams only a single directed electron beam source, the so-called radial area beam, is used which is arranged in the centre axis of a tube-shaped exhaust gas channel and beams in at least two directions radially towards the outer wall of the exhaust gas cahnnel. The radial area beam, E (Figs 1 and 2) is sited coaxially in the exhaust gas channel 10 in a cylinder-shaped device and has a minimum of two, but preferably four or more electron emission apertures 6. Although this arrangement of the electron beam source E, results in an unhomogeneous dosage distribution in the exhaust gas channel, this is unimportant as in irradiation from outside to the interior with two beams opposite each other a turbulent exhaust gas flow is also used.

The distance between the electron emission apertures 6 and the outer wall 10 of the exhaust gas channel depends on the maximum potential of the beam. It must always be ensured that the maximum range of the electron beams is not greater than the distance between the aperture and the outer wall as otherwise the pipe wall would heat up leading to reduced efficiency in the irradiation device.

The X-ray shield of the radial area beam is of a particularly simple form according to the present invention. The following are possible:

(a) With the arrangement of the radial area in an open horizontal exhaust gas channel (figure 3) the channel is clad directly with lead sheet. To break the X-rays spreading out in the channel, the channel is bent twice, before and after the irradiation zone. These bends are also clad with lead, the manufacture of this lead cladding being simple as flat surfaces are involved.

The electrical and mechanical supply 12 to the electron beam is via one facing edge of the bent exhaust gas channel. This facing edge also serves as a servicing opening for the radial area beam. The radial area beam can be removed from the exhaust gas channel via this point for servicing.

(b) With the arrangement of the radial area beam in a vertical exhaust gas channel (figure 4) there is the possibility of the channel running through the earth which can then also act as an X-ray shield. For servicing purposes, the radial area beam is removed from the exhaust gas channel through its face edge.

The radial area beam consists of components known in electron beam technology, the area beam having a long service life and the electron emission aperture opposite the cathode system being designed in the same way as described in the applicant's prior application.

The physical basis for optimising the radial area beam and the exhaust gas channel will be described in th following.

One-step electron accelerators are today manufactured with a beam potential of from 150 kV to 300 kV. The beam potential is limited at the lower end of the scale by energy losses in the electron emission aperture and at the upper end of the scale by the high potential strength of the one-step distance of accel eration.

The following calculations relate to a *theoretical* beam potential of 600 kV. In industry today beam potentials of 300 kV are reached. If the beam potential remains below this figure of 300 kV, the operational safety of the irradiation device is increased.

When irradiating exhaust gas it must be ensured that the flow in the exhaust gas channel lies between 15 and 20 m/s, and in special cases 30 m/s.

The calculations are based on a 500 MW_{el}, power station, which corresponds to an exhaust gas output of 1,500,000 Nm^3/h (cubic metres per hour at normal or standard pressure and temperature). At an exhaust gas temperature of approx 80–100°C this results in 2 million m^3/h or 555 m^3/s of exhaust gas. 1 m^3 of exhaust gas is taken to weigh 1 kg.

Dose formula: 1 Mrd = 10 kGy = 10 kJ/kg = 10 kW. s/kg

Thus the decontamination of a 500 MW_{el} power station at a dosage of 1 Mrd requires an effective irradiation of 5,500 kW_{eff}.

An industrially manufactured electron beam contains the following principal components:
Cathode, pre-acceleration distance, post-acceleration distance, electron emission apertures.
An electron emission aperture can be 200 cm in l ngth with an operating width of 22 cm.
The aperture loading is 0.15 mA/cm^2.
The transmission of a supported electron emission aperture is $\eta = 50\%$.
The electron current of an aperture is thus 660 mA corresponding to 330 mA_{eff}.

In the following, *two electron area beams*, each equipped with two electron emission apertures and which irradiate opposite each other in a right-angled exhaust gas channel (figure 5), are compared with *one radial area beam* arranged in a tube-shaped exhaust gas channel and equipped with four electron emission

apertures.

The comparison is intended to result in optimisation in both cases as regards the number of electron beams, beam potential and number of exhaust gas channels.

A further parameter is the number of electron beams for a dose of 1 Mrd as well as the number of electron beams for a does of 0.5 Mrd.

New information obtained from experimental work has shown that in order for decontamination to take place, an electron beam dose of down to 0.3 Mrd could be possible to a traditional desulphurisation stage, Such low irradiation doses are of course extremely important for the efficiency of the process.

Figure 6 (according to Bailey) serves to calculate the maximum range of the electron geam as a function of the beam potential. The lower curve takes into account the passage of the electrons through a 15 mm titanium sheet, corresponding to a surface weight of 6.75 mg/cm^2.

1. First comparison of ideal electron beams with ideal exhaust gas channel.

Two electron area beams arranged opposite each other in a right angled exhaust gas channel,

The columns in the following table 1, refer to:

kV	Beam potential of the electrons in kV
kW_{eff}	Total effective electron beam output of the 4 electron emssion apertures
n_1	Number of ideal electron beams required at a dosage of 1 Mrd.
$N_{o.5}$	Number of ideal electron beams required at a dosage of 0.5 Mrd.
r_o in mg/cm^2	maximum range of the electron beams in exhaust gas with a density of 1 kg/m^3
$d_{o.7}$ in m	Depth of the exhaust gas channel with two-sided irradiation and overlapping of the bell-shaped ionisation curves (figure 7) $d_{o.7}$ consists of 2 times $r_o.0.7 = r_o. 1.4$ and takes into account the degree of overlapping
$d_{o.8}$ in m	As $d_{o.7}$ but lower overlapping factor, r_o . 1.6
$F_1.4$ in m^2	Cross-section area of exhaust gas channel in m^2 for $d_{o.7}$
$F_{1.6}$ in m^2	As $F_{1.4}$ but for $d_{o.8}$
m^3.s^{-1}	Throughput in m^3 . s^{-1} per exhaust gas channel cross-section for channel depth 1.4 (r_o . 1.4)
N_{15} 1.4	Number of exhaust gas channels for decontamination of a 500 MW_{el} power station, for 15 m/s exhaust gas speed and channel depth 1.4 (r_o . 1.4)

These calculations were carried out for 15 m/s and 30 m/s exhaust gas speed and a channel depth factor of 0.7 and 0.8 (r_o . 1.4 and r_o . 1.6).

The results of these calculations are set out in figure 8.

Ideal values for the number of electron beams and exhaust gas channels are found at the intersection of the curves n_1, for ideal electron beam 1 Mrd or $n_{o.5}$ for ideal electron beam for a dosage of 0.5 Mrd, with curves $n_{16\ 1.4}$ to $N_{30\ 1.6}$ for the ideal number of exhaust gas channels.

It should be noted that the number of ideal electron beams always consists of *two electron area beams opposite each other.*

2. Second comparison of ideal electron beams with ideal exhaust gas channel.

A radial area beam, concentric in circular exhaust gas channel.

The individual columns in the following table 2 refer to:

kV	Beam potential of the electrons in kV
kW_{eff}	Total effective electron beam output of the radial area beam with a total of 4 electron emission apertures
n_1	Number of ideal electron beams required for decontamination at a dosage of 1 Mrd
$N_{o.5}$	Number of ideal electron beams required for decontamination at a dosage of 0.5 Mrd
r_o in mg/cm^2	Maximum range of electron beams in exhaust gas with density of 1 kg/m^3
d_1 in m	Diameter of the almost circular radial area beam
d_2 in m	Diameter of the exhaust gas channel taking into account the maximum range r_o of the electron beams
F_1 in m^2	Cross-sectional area of the radial area beam
F_2 in m^2	Cross-sectional area of the exhaust gas channel with radial area beam
<F in m^2	F_2 minus F_1 and the thus resulting cross-sectional area of the exhaust gas channel
m^3 . s^{-1}	Throughput in m^3. s^{-1} per exhaust gas channel for various exhaust gas speeds
n_{15}	Number of exhaust gas channels for the decontamination of a 500 MW_{el} power station at 15 m/s exhaust gas speed.

These calculations were carried out for 15 m/s, 20 m/s and 30 m/s exhaust gas speeds.

The results of these calculations are set out in figure 9.

The ideal values for the number of electron beams and exhaust gas channels are found at the intersection of the curves n_1, for ideal electron beams 1 Mrd or $n_{o.5}$ for ideal el ctr n beams with an irradiation dose of 0.5 Mtd, with curves n_{15} to n_{30} for the number of ideal exhaust gas channels.

Discussion of comparisons 1 and 2:

(a) The radial area beam is of a simpler design as it only requires a high potential device, a vacuum

element and a control d vice.

(b) The radial area beam requires, depending on exhaust gas spe d and dose requirements, a beam potential of max 300 kV, and with higher exhaust gas speeds and lower irradiation doses even lower beam potentials, which increases the operational security.

(c) As the exhaust gas flow is turbulent even with electron area beams opposing one another, one-sider irradiation of the exhaust gas with a radial area beam should not be disadvantageous.

(d) X-Ray shielding for the radial area beam is solved optimally.

(e) When two electron area beams arranged opposite each other are used, ie two-sided irradiation, the ideal beam potentials are, depending on exhaust gas speed and necessary dosage, between 300 and 600 kV. These beam potentials are simply not attainable.

(f) Two electron area beams must always be used per exhaust gas channel in two-sided irradiation.

(g) Two-sided irradiation complicates the shielding possibilities.

TABLE 1

Comparison: Number of ideal beams with number of ideal gas channels as a function of beam potential.
Power Station: 500 MW_{el} ≙ 1.500.000 Nm^3/h = 2.00.000 m^3/h = 555 mV/g = 555 kg/s
≙ 5.550 KW_{eff} irradiation
Beams: 200 cm long. Each has 2 × 22cm wide windows. 0,15 mA/cm^2, η = 50%
Arrangement of 2 double beams opposite each other, 2.640 mA_{gs}/1320 mA_{eff}

kV	kW_{eff}	n_1 Number of beams required for a dose of 1 Mrd	$n_{0,5}$ 0,5 Mrd	r_o (mg/cm^2)	$d_{0,7}$ $2.r_o.0,7$	$d_{0,8}$ $2.r_o.0,8$	$F_{1,4}$ for (m) $d_{0,7}$	$F_{1,6}$ for (m) $d_{0,8}$
150	198	28	14	23	0,322	0,368	0,644	0,736
180	237,6	23,3	11,6	33	0,462	0,528	0,924	1,056
210	277,2	20	10	44	0,616	0,704	1,232	1,408
230	303,6	18,3	9,1	52	0,728	0,832	1,456	1,664
250	330	16,8	8,4	60	0,84	0,96	1,68	1,92
280	370	15	7,5	73	1,022	1,168	2,044	2,336
300	396	14	7	83	1,162	1,328	2,324	2,656
400	528	10,5	5,2	138	1,932	2,208	3,864	4,416
500	660	8,4	4,2	195	2,73	3,12	5,46	6,24
600	792	7	3,5	260	3,64	4,16	7,28	8,32

$m^3.s^{-1}_{1,4}$ 15 m/s	n_{15} Number of channels	$m^3.s^{-1}_{1,6}$ 15 m/s	n_{15} 1,6 Number of channels	$m^3.s^{-1}_{1,4}$ 20 m/s	n_{20} 1,4 Number of channels	$m^3.s^{-1}_{1,6}$ 20 m/s	n_{20} 1,6 Number of channels	$m^3.s^{-1}_{1,4}$ 30 m/s	n_{30} 1,4 Number of channels	$m^3.s^{-1}_{1,6}$ 30 m/s	n_{30} 1,6 Number of channels
9,6	57,4	11,0	50,3	12,8	43	14,7	37,7	19,3	28,7	22,0	25,1
13,8	40	15,8	35	18,4	30	21,1	26,3	27,7	20	31,6	17,5
18,4	30	21,1	26,3	24,6	22,5	28,1	19,7	36,9	15	42,2	13,1
21,8	25,4	24,9	22,2	29,1	19	33,2	16,7	43,6	12,7	49,9	11,1
25,2	22	28,8	19,3	33,6	16,5	38,4	14,4	50,4	11	57,6	9,6
30,6	18,1	35,0	15,8	40,8	13,6	46,7	11,9	61,3	9	70,0	8
34,8	15,9	39,8	13,9	46,4	11,9	53,1	10,4	69,7	8	79,6	7
57,9	9,6	66,2	8,4	77,2	7,2	88,3	6,3	115,9	4,8	132,4	4,2
81,9	6,8	93,6	5,9	109,2	5,1	124,8	4,4	163,8	3,4	187,2	3
109,2	5,1	124,8	4,4	145,6	3,8	166,4	3,3	218,4	2,5	249,6	2,2

TABLE 2

Comparison: Number of ideal beams with number of ideal exhaust gas channels as function of the beam potential.
Power Station: 500 MW_{el} ≙ 1.500.000 Nm^3/h = 2.000.000mm^3/h = 555 m^3/g = 555 kg Rauchgas
≙ 5.500 kW_{eff} irradiation
Beams: Radial area electron beamsn, 200 cm long. Each beam has 4 × 22 cm wide windows.
0,15 mA/cm^2, η = 50%, 2640 mA_{gs}/1320 mA_{eff}
Cylindrical exhaust gas channel.

kV	kW_{eff}	n_1 Number of beams required for a dose of 1 Mrd	$n_{0,5}$ 0,5 Mrd	r_o (mg/cm^2)	d_1 (m)	d_2 (m^2)	F_1 (m^2)	F_2 (m^2)	F (m^2) F_2 minus F_1
150	198	28	14	23	1,2	1,66	1,13	2,16	1,03
180	237,6	23,3	11,6	33	1,2	1,86	1,13	2,71	1,58
210	277,2	20	10	44	1,2	2,08	1,13	3,39	2,26
230	303,6	18,3	9,1	52	1,2	2,24	1,13	3,94	2,80
250	330	16,8	8,4	60	1,2	2,4	1,13	4,52	3,39
280	370	15	7,5	73	1,2	2,66	1,13	5,55	4,42
300	396	14	7	83	1,2	2,86	1,13	6,42	5,29
400	528	10,5	5,2	138	1,2	3,96	1,13	12,31	11,18
500	660	8,4	4,2	195	1,2	5,1	1,13	20,4	19,28
600	792	7	3,5	260	1,2	6,4	1,13	32,15	31,02

$m^3.s^{-1}$ 15m/s	n^{15} Number of channels	$m^3.s^{-1}$ 30 m/s	n_{20} Number of channels	$m^3.s^{-1}$ 30 m/s	n_{30} Number of channels
15,4	35,9	20,6	26,9	30,9	18
23,7	23,4	31,6	17,6	47,4	11,7
33,9	16,4	45,2	12,3	67,8	8,2
42,0	13,2	56,0	9,9	84,0	6,6
50,8	10,9	67,8	8,2	101,7	5,4
66,3	8,4	88,4	6,3	132,6	4,2
79,3	7	105,8	5,2	158,7	3,5
167,7	3,3	223,6	2,5	335,4	1,6
289,2	1,9	385,6	1,4	578,4	0,95
465,3	1,2	620,4	0,9	930,6	0,6

CLAIMS

1. Apparatus for the desulphurisation and denitration of exhaust gases by electron irradiation of the exhaust gases to which ammonia has been added before irradiation, comprising an exhaust gas channel and a minimum of one low energy electron beam source with an electron beam potential of 15–300 keV in which the electron beam source is arranged con-centrically and coaxially in the exhaust gas channel and has at least two electron emission apertures.

2. The apparatus of claim 1, in which the electron beam source has four or more electron emission apertures.

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